for compensation legislation; and (5) EH Policy Integration project to effectively communicate the EH gaseous diffusion plant projects to key stakeholders and across the DOE organization [2].

This report addresses activities at the Savannah River Site conducted in support of the Recycled Uranium Mass Balance Project (item 2 above).

1.2 Purpose and Scope

The purpose of this report is to quantitatively estimate the flow and characteristics of recycled uranium received, handled, and shipped from the Savannah River Site. This quantitative information will enable an assessment of the potential for worker exposure to radioactivity in the uranium stream, including that from technetium-99, neptunium-237, and plutonium.

This report includes all recycled uranium processed at SRS since site startup, covering the period from the early days of production to March 31, 1999.

1.3 Project Implementation Strategy

The recycle uranium mass balance project at the SRS was conducted by an inter-disciplinary site team composed of contractor and DOE-SR personnel, including personnel knowledgeable in nuclear material control and accountability, radiological health protection, process operations, analytical methods, records management, and environmental management. The team's focus was on identifying recycle uranium flows into and out of the site, the contaminant constituents present in the material shipped and received, and the impact of those contaminants on worker safety/health and the environment.

Research of historical records formed the basis for the team's conduct of this project. Project results were validated through comparison of those records with known site operating history, and with records from receiving/shipping sites throughout the complex.

2.0 Site Historical Overview

2.1 Site Description

The Savannah River Site encompasses approximately 310 square miles including portions of Aiken, Barnwell, and Allendale Counties in South Carolina. The site is situated adjacent to the Savannah River approximately 12 miles south of Aiken, South Carolina. It consists of sixteen (16) distinct areas of operation as depicted in Figure 1-3. The site was established, by the U. S. Atomic Energy Commission (AEC) in 1950, to produce plutonium and tritium for national defense and additional special nuclear materials for other government uses and for civilian purposes.

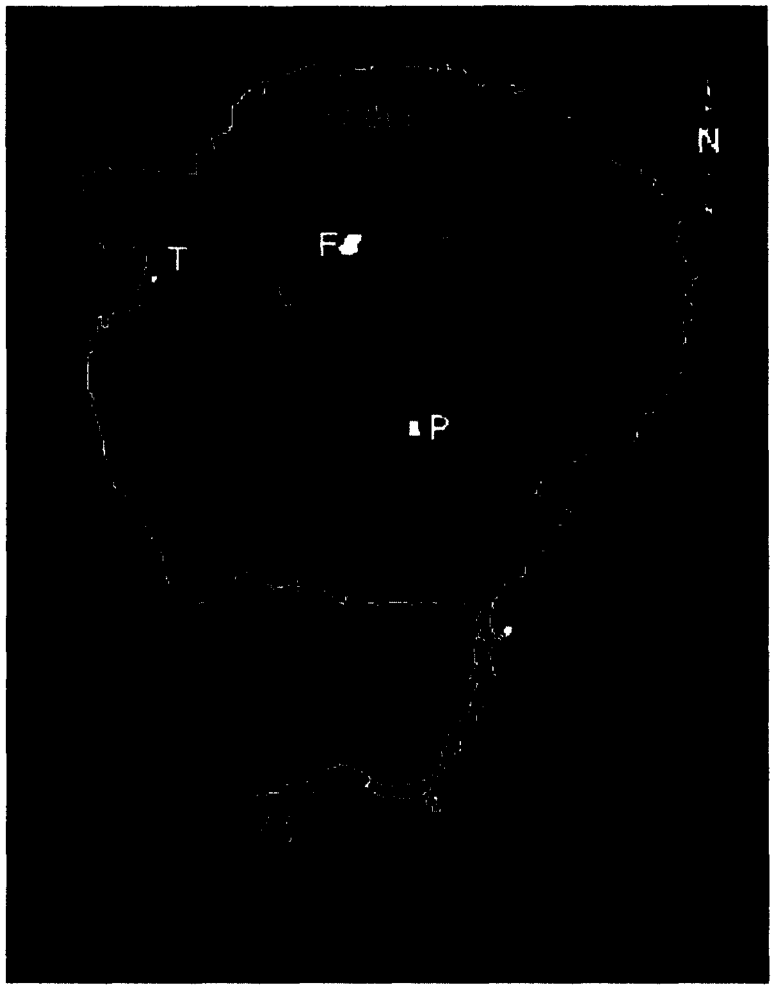
On July 25, 1950 President Harry S. Truman sent a letter to E. I. DuPont De Nemours Company President, Crawford Greenewalt requesting that DuPont undertake a new project for the Atomic Energy Commission. In response to the Truman letter and a directive to the AEC, the DuPont Company and the Atomic Energy Commission negotiated a contract whereby DuPont would design, construct, and operate what was to become the Savannah River Plant (SRP).

On November 22, 1950 the AEC approved the present site and purchased the land for approximately $19 million. By February 1, 1951, construction had begun. The Heavy Water Plant was the first facility in operation, starting up August 17, 1952, with the first of five production reactors beginning operation on December 28, 1953. All five reactors had become operational by March 1955 [3].

Plant oversight and regulation was provided by the AEC until it was disbanded by the Energy Reorganization Act of 1974. Plant oversight and regulation was transferred in 1975 to two new agencies—the Energy Research and Development Administration (ERDA) overseeing government operations and the Nuclear Regulatory Commission (NRC) overseeing commercial operations. In 1977, ERDA evolved into the Department of Energy (DOE), which has overseen site activities since that time.

DuPont operated SRP until March 31, 1989, and on April 1, 1989 the Westinghouse Savannah River Company (WSRC) became the prime operating contractor, at which time SRP became SRS. Beginning October 1, 1996, the site was operated under a new contract by an integrated team led by WSRC. Under the new contract, WSRC is responsible for SRS's nuclear facility operations, Savannah River Technology Center (SRTC), Environment, Safety, Health, and Quality Assurance, and all the site's administrative functions. Bechtel Savannah River, Inc. is responsible for Environmental Restoration, Project Management, Engineering, and Construction activities; Babcock and Wilcox Savannah River Company is responsible for Decontamination and Decommissioning; and British Nuclear Fuels Savannah River Corporation is responsible for the site's Solid Waste Program [4].

FIGURE 1-3
The Savannah River Site



2.2 Key Uranium Processing Facilities

This section provides a brief overview of the SRS processes used to produce nuclear materials. A more detailed discussion of the processes, which involved the handling of recycled uranium with potential for personnel exposure and environmental impact will follow. SRS was constructed to produce basic materials used in nuclear weapons, primarily tritium and plutonium-239. Five reactors were built to produce these materials by irradiating target materials with neutrons. Support facilities also were built, including two chemical separation plants, a heavy water extraction plant, a nuclear fuel/target fabrication facility, and waste management facilities.

The production process began with the manufacture of fuel and target assemblies produced from a variety of nuclear and other materials such as enriched uranium and aluminum. The assemblies were transported to the reactors, where they were irradiated to produce the desired products. The irradiated target assemblies and spent fuel assemblies were moved to the chemical separation facilities where the desired products were separated and waste products were processed. After refinement, nuclear materials were shipped to other DOE sites for use in the nuclear weapons program or the NASA deep space program.

Changing world conditions have impacted traditional SRS missions, resulting in downsizing of the site's defense mission and workforce. All five reactors are now shut down, however, recycling and reloading of tritium to maintain the nation's supply of nuclear weapons is a continuing site mission. The current SRS mission is to serve the national interest by ensuring that program operations, and resources are managed in a safe, open, and cost-effective manner which;

(1) supports current and future national security requirements;

(2) reduces the global nuclear proliferation danger;

(3) protects and restores the environment while managing waste and nuclear materials;

(4) conducts mission-supportive research and technology development.

The site's priorities today involve; (1) cleaning up waste sites by removing hazardous substances or by stabilizing, containing, or treating substances so that they do not affect human health or the environment (environmental restoration); (2) managing newly generated waste and waste that is a legacy from the production of nuclear materials (waste management); (3) "disposition" of excess facilities that are no longer needed to produce or process nuclear materials; and (4) managing nuclear materials in an effort to curb the spread of nuclear weapons [4].

FIGURE 1-4
SRS Fuel Fabrication Facility Building 321-M

2.2.1 Uranium Fabrication

2.2.1.1 Plant Description

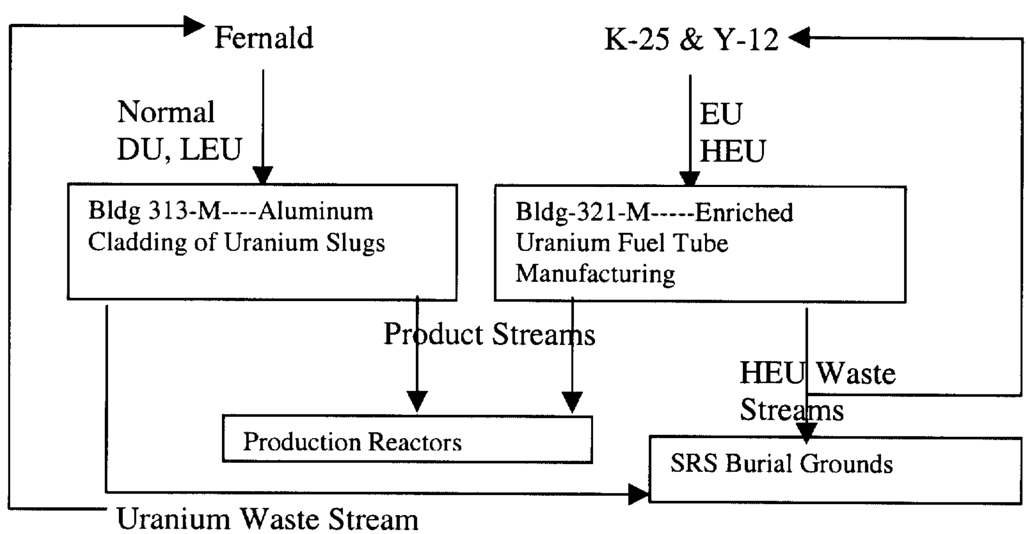
Uranium materials generally entered the site through the Fuel and Target Fabrication Facilities located in the 300 Area or Raw Materials Area, later called Reactor Materials Area (Figure 1-4 above shows Bldg.321-M, Fuel Fabrication Facility prior to 1997). The uranium forms handled in Fuel and Target Fabrication Facilities were naturally radioactive. Additional radioactive constituents, resulting from recycling of uranium, were also present but in insufficient concentrations to create a radiological hazard greater than that posed by the uranium itself. None of the processing operations in these facilities had the capability to concentrate the radioactive constituents. A brief description of those processes is provided here. At the Fabrication Facilities, materials including lithium, depleted uranium, enriched uranium, normal uranium, and aluminum went through various metal working operations such as casting, machining, plating, extrusion, and welding. Depleted normal, and slightly enriched uranium (LEU) fuel cores were received from the Feed Materials Production Center

(FMPC) at Fernald, Ohio, for treatment and canning at the 313-M building. Highly enriched uranium metal for driver fuel was received from the Y-12 Plant at Oak Ridge, TN, into the 321-M building, where it was alloyed with aluminum. The highly enriched uranium and aluminum (HEU-Al) billets were extruded into fuel tubes for use with targets of lithium or depleted uranium.

Tubular fuel elements were manufactured in the 321-M building by co-extrusion of a composite billet. In this process, the tube is formed and the core is simultaneously clad with aluminum. Cores consisted of fuel material dispersed in an aluminum matrix. Most elements manufactured in the 321-M building contained highly enriched uranium cores. After extrusion, tubes were chemically cleaned to remove graphite and lead-oil extrusion lubricant. They were successively treated with hot perchloroethylene, sodium hydroxide, and nitric acid; intermediate and final water rinses were also used.

In the 313-M building, target and fuel elements from natural, depleted, or slightly enriched uranium were bonded in aluminum cans by several techniques. From 1968, only depleted uranium cores were used in conjunction with highly enriched drivers, with the exception of some experimental cores containing 1.1 percent U-235 processed in 1982. The depleted uranium cores were cleaned at SRS with boiling perchloroethylene and hot nitric acid, anodically etched, and then electroplated to form a 0.3-mil-thick nickel layer. The nickel plating was required to assure a good bond with the aluminum can and protect the uranium from oxidation during heating. The nickel-plated core was loaded into an aluminum can, capped, preheated, and pressed through a die to size the can onto the core. The canned elements were then cleaned, welded, inspected, and tested [5].

2.2.1.2 Material Flowsheet



2.2.1.3 Material Feed Specifications

Enriched Uranium Specification (Feed/Product)

No one established specification for transuranics in enriched uranium material shipped from SRS was found. Located were proposed specifications from Fernald and Y-12, but no evidence was found that these were ever officially incorporated into an SRS specification [19], [20]. Shipping records indicate that the product specification for slightly enriched uranium shipped from SRS was expressed as 1 Pu alpha d/m/700 U alpha d/m or as 3000 Np + Pu alpha d/m/gram U, although no official plant specification was found to confirm these values. Recycled Enriched Uranium Specification # A48177-321M, Rev. 0, dated 4/6/90 provides the most current specification for radioactive contaminants permissible in material received at SRS [6].

The specification requires that the total gamma activity from radioisotopes of fission products and induced activities shall not exceed 0.3 micro-curies/gram of uranium. It further requires that the gamma activity from individual radio nuclides shall not exceed the following:

Radionuclide	Maximum gamma Activity, micro-curies/gU
Cerium	0.05
Ruthenium	0.05
Cesium	0.05
Zirconium-Niobium	0.01
Any other individual radio-nuclide	0.05

The specification requires that the total alpha activity from Neptunium and plutonium shall not exceed 0.1 micro-curies/gram of uranium.

These specifications are based on the results of early experience gained handling recycled enriched uranium at SRS [6].

Uranium Oxide Specification (Feed/Product)

SRS specifications for depleted UO3 returned to the Gaseous Diffusion Plants (GDPs) for feed were based on gamma and beta activity as a percent of aged normal uranium for fission products plus U-237. The maximum gamma activity for one carload of material was 300 percent, and the ten carload average maximum was limited to 100 percent of aged normal uranium. The beta activity for one carload was a maximum 100 percent of normal aged uranium.

The specification for plutonium that could be present in the UO3 was 10 ppb. This specification was provided to SRS in an August 1954 letter KLI-3091, from D. M. Lang, Technical Division Superintendent of Oak Ridge K-25 Plant to L. C. Perry, Superintendent Separation Technology Section of SRS. This specification has remained constant over the years [7], [8].

2.2.1.4 Operating History

Fuel and target fabrication operations at the SRS began in 1952. The fuel elements and the neutron-absorbing lithium-aluminum alloy cylinders for the control rods were needed for the reactors as soon as the reactors were filled with heavy water. Therefore, construction of the 300-M Area had begun at about the same time as that of the heavy water area.

Employee safety was of prime concern due to the many hazards involved in 300-M area operations. These hazards included chemical cleaning lines with strong acids and caustics, processes with extremely high operating temperatures, criticality concerns, and large/powerful presses and lathes. The plants were operated under strict administrative and engineering controls.

Over one-quarter-million slugs, housings, control rods, fuel tubes, and target tubes were produced annually by about 300 workers, during periods of highest production of plant operations. Reactor charge design changes necessitated numerous rapid changes in component design and manufacturing techniques by 300-M area personnel 2-3 years prior to when the components would be needed by reactors. Cladding technologies were greatly improved, as were methods for inspection and testing, particularly the final test of each fuel and target assembly for nuclear reactivity. On-line computer surveillance and guidance enhanced the safety and efficiency, for fabrication of enriched uranium fuel tubes. The co-extruded aluminum-clad enriched-uranium fuel tubes were remarkable accomplishments of metallurgical technology [3].

2.2.1.5 Current Status

Fuel and target processing efforts ceased in the fabrication facilities in the early 1990s. Approximately 2,600 metric tons of depleted uranium and 35 metric tons of normal uranium remain in storage in M-Area facilities. An estimated 2 kg of enriched uranium remains in Building 321-M as residue holdup in equipment and ventilation ductwork. Equipment that could be salvaged for use by commercial operations has been sold and/or released as part of the site's technology transfer program. The buildings have been turned over to the site's Facilities Disposition Division.

2.2.2 Uranium Irradiation

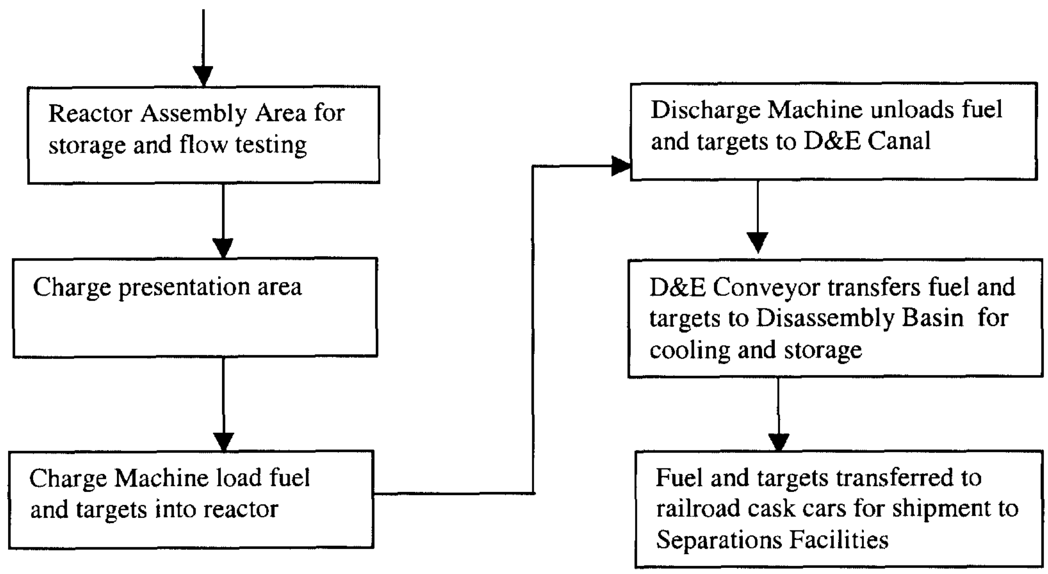
2.2.2.1 Plant Description

Finished fuel and target assemblies were transported to site reactors and subjected to an irradiation cycle where plutonium and tritium were produced for nuclear weapons and small quantities of other radioisotopes were produced primarily for non-military purposes. Handling of the uranium prior to irradiation did not subject workers to un-due radiation hazards, and handling of uranium after irradiation was done remotely to protect workers from radiation hazards. Since little opportunity existed for internal uptake of uranium and its associated constituents by reactor workers, only a brief description of the fuel handling and reactor loading operations will be discussed in this section.

Fresh fuel was received and stored in the reactor assembly area where it was placed on racks and hangers in a configuration that prevented criticality. For driver assemblies, a neutron poison was added to the storage racks (e.g., borated concrete). The reactor room was equipped with a charge machine, discharge machine, and an irradiated component conveyor. Charge and discharge machines were used to remotely load and unload fuel and target assemblies from the reactor. These operations were conducted from the crane control room located adjacent to the reactor room. The conveyor was located in a water-filled canal, which connected the reactor room with the irradiated assembly storage basin. The conveyor received irradiated assemblies from the discharge machine and passed them under the wall of the reactor room. Irradiated assemblies were stored vertically in the water-filled storage basin until their decay heat was low enough to permit shipment to the separation facilities [5].

2.2.2.2 Material Flowsheet

Fuel and Target Assemblies from 300-M Area



2.2.2.3 Feed Specification

Same as that stated in section 2.2.1.3 above.

2.2.2.4 Product Specification

Same as that stated in section 2.2.1.3 above.

2.2.2.5 Operating History

Initial design of Savannah River Site reactor systems began in August 1950, and fabrication of the first reactor by New York Shipbuilding Company began in September 1951. All five reactors (R, P, L, K, C) were fabricated between September 1951 and May 1954 and were turned over to Savannah River Operations between April 1953 and September 1954. The first reactor placed in service was R-Reactor, which started up in December 1953. The last, C-Reactor was started up in March 1955.

Through the years SRS reactors utilized three basic charge designs:

1) Uniform Core – Usually up to 600 assemblies (588 in C-Reactor) of the same type. Each assembly could contain both fissile and target material.

2) Mixed-Lattice Core – These charges contained a large number of driver and target assemblies intermixed in a ratio of 3:3, 4:2, or 5:1 arranged with six assemblies surrounding each control rod position.

3) Small Core – Reduced portion of the reactor tank (both radially and axially with height of active core typically about equal to diameter) is used to attain a high neutron flux and high power density. Typically, all heavy water coolant flow would be directed to between 100 to 400 lattice positions, occupied by either an essentially uniform or a mixed-lattice core. The site had run three small core campaigns, which included a Californium-252 campaign by 1981.

SRS charges were designed to produce tritium, plutonium-239, and other radionuclides. Initial criticality in R-Reactor in 1953 used a natural uranium charge for production of plutonium. Over the years mixed cores containing assemblies with a variety of uranium enrichments have been irradiated in SRS reactors. However, since 1968 SRS reactors have used only mixed cores of depleted uranium targets and high burn-up, highly enriched drivers in plutonium production campaigns. Since about 1957, a uniform lattice (HEU drivers with integral Li-Al targets) has been the charge design of choice for the production of tritium at SRS [5].

Of constant concern to management, and to reactor engineers and scientists, were the risks of reactor operation and ways to reduce the risks and mitigate the effects of an accident should one occur. Management recognized that as power levels were raised, so too were the consequences of potential accidents. Efforts to reduce the risks, prevent and mitigate the effects of accidents were continuous, and were increasingly effective as understanding of the physics of heavy-water reactors increased and deepened.

Safety was greatly enhanced by advancements made in electronics and computers. These new technologies were quickly adopted and adapted, as they became available. SRS was at the forefront of the nuclear industry in the development and applications of computers to reactor surveillance and operating control.

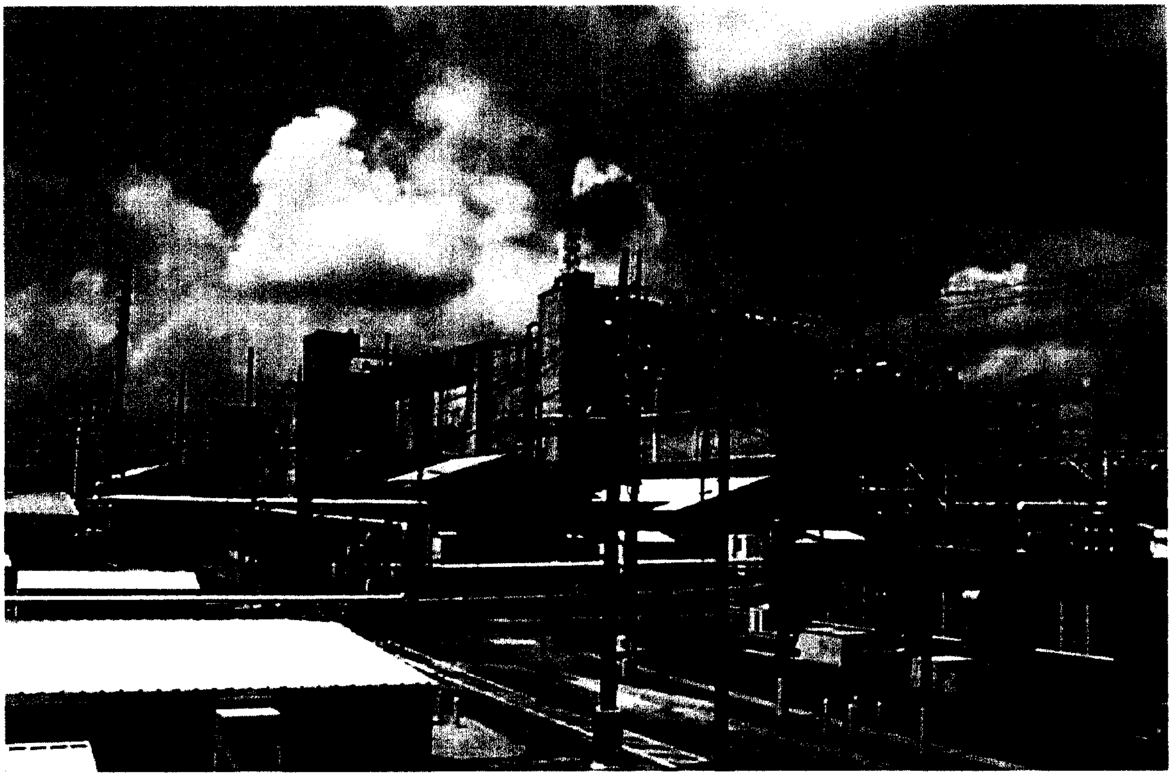
Emphasis on reactor and personnel safety is evident in the record of safety of SRS reactors. These reactors were operated for over 40 years and never suffered an accident that seriously damaged reactor equipment, or seriously injured an employee [3].

2.2.2.6 Current Status

For all intents and purposes reactors operations ceased at the SRS in 1988. Though efforts were made to restart K-Reactor in the early 1990s as the cold-war was winding down, these efforts were abandoned with the end of the cold-war.

2.2.3 Uranium Separation

FIGURE 1-5 – F-Area Canyon Facility



2.2.3.1 Plant Description

Irradiated assemblies from the reactors are transported to two large chemical processing plants (one in the 200-F Area and one in 200-H Area). Operations in these areas presents the greatest potential for worker internal assimilation of uranium and uranium by-products, and will be described in some detail. The F-Area plant recovers and separates Pu-239, Np-237, and U-238 from irradiated natural or depleted uranium targets. The H-Area plant recovers uranium isotopes, Np-237, and Pu-238 from irradiated enriched uranium fuel assemblies and converts the isotopes into a desired form for shipment. Both canyons remain in operation today and are being used to stabilize material from this site and from around the DOE Complex.

Casks containing material for processing are delivered by a plant-operated railroad to the hot canyon. In both F and H chemical processing plants, the first step is to dissolve the fuel, a process that liberates volatile fission products and generates solutions with high concentrations of radioactivity. The initial separation yields solutions of plutonium, uranium, or neptunium product, and a high-activity liquid waste containing non-volatile fission products. After the fission products are removed sufficiently from the product solutions, further processing can be done in unshielded areas, where product (e.g., plutonium and uranium) may be converted from solution form to solids. The A-Line Facility in F-Area converted recovered U-238 nitrate solutions to uranium trioxide powder. In H-Area, a uranium solution trailer-loading station is sometimes referred to as H-Area A-Line. H Canyon shipped liquid uranyl nitrate to the Y-12 Plant at Oak Ridge for conversion to metal until the late 1980's.

In F-Area Canyon building, the PUREX solvent extraction process is used to recover Pu-239 from irradiated uranium targets. Fuel processing begins with dissolution of the fuel. In the case of normal aluminum-clad, metallic uranium fuel, the aluminum cladding is dissolved in sodium hydroxide-sodium nitrate solution, with evolution of ammonia and hydrogen. De-clad fuel is dissolved in nitric acid, following the removal of the de-cladding solution with contained aluminum.

In the first solvent extraction cycle, uranium and plutonium are extracted away from the remaining fission products with tributyl phosphate in a hydrocarbon dilutent and then separated from each other. The low enriched uranium and plutonium are further purified by separate, second solvent extraction cycles. The final low enriched uranium product solution (uranyl nitrate) is concentrated and sent outside the canyon for conversion to uranium trioxide in the A-Line facility. The uranium trioxide powder is stored onsite. In peak production years during the Cold War, F-Area generated 2000, to 3000 drums of uranium trioxide powder per year. Currently, approximately 20,000 metric tons of this material is in long-term storage at the site.

In the warm canyon of the 221-F building, neptunium and residual plutonium were recovered by ion exchange from the fission product waste solution from solvent extraction. Neptunium was further purified by ion exchange, and the final product solution was sent to the H-Area neptunium line or HB-Line. The HB-Line converted neptunium-237 and plutonium-238 from both chemical separation areas to powder. The plutonium was sent to a separate operation, the JB-Line, atop the 221-F canyon building where the plutonium was concentrated by ion exchange, precipitated as fluoride, and, with some further treatments, reduced to metal buttons. The metal buttons were shipped to Rocky Flats for manufacture into plutonium components of nuclear weapons.

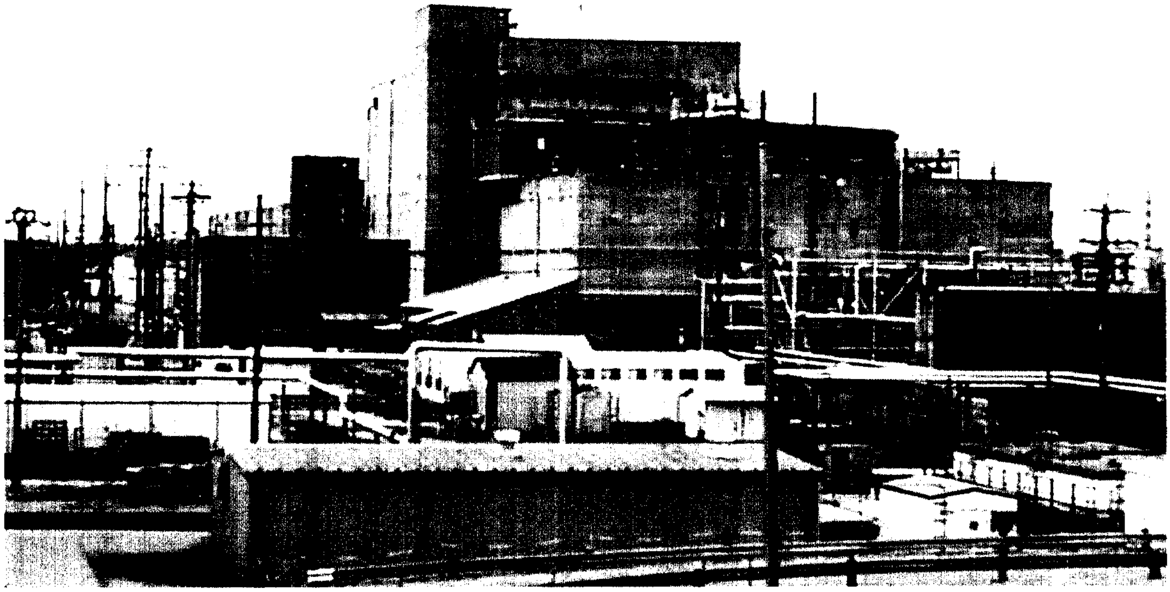


FIGURE 1-6 – H-Area Canyon Facility

In the 221-H building the HM-process ("H modified" PUREX process) is used to separate uranium and neptunium, or plutonium from irradiated fuels containing from 1.1 percent to 94 percent uranium-235. Since 1959 H-Canyon has been devoted primarily to the recovery of enriched uranium from SRP fuel. Enriched uranium is also recovered from fuel of domestic research reactors and from foreign research reactors fueled with material produced or enriched by the United States.

The H-Canyon facility dissolves fuels with a variety of claddings. Aluminum clad fuels are dissolved directly in nitric acid; zirconium or stainless cladding is first removed in an electrolytic dissolver, with nitric acid electrolyte, and then the core material is dissolved chemically in nitric acid. The solvent extraction operations are similar to those of the PUREX system. Some chemical processes are different because of differences in the fuels. Uranium and neptunium are extracted from the fission products and separated from each other in the first cycle; then each is further purified by two separate cycles of solvent extraction. The highly enriched uranium product solution was not concentrated further, but was transferred out of the building, loaded into tank trucks, and sent to the Y-12 Plant at Oak Ridge for conversion into uranium metal to be recycled as driver fuel for the SRS production reactors. Low enriched uranium (2 to 10 percent U-235), from the processing of fuel from government owned test reactors, was recovered as uranyl nitrate and shipped to the Feed Material Production Center (Fernald Plant) for conversion into UF-6 feed for the enrichment plants. The neptunium product solution was transferred to a separate area in the building for separate purification.

In a separate process system in the shielded H canyon, irradiated neptunium-237 targets were dissolved in nitric acid, and plutonium-238 and neptunium are separated from fission products and each other by a series of anion exchange resin columns. The product solutions of plutonium-238 and neptunium-237 are transferred to the finishing area, the HB-Line, where the two are concentrated, precipitated as oxalate, and calcined to oxides. The plutonium oxide is packaged for shipment offsite, and the neptunium oxide is sent to the metallurgical building (235-F) for re-fabrication into billets to be made into reactor neptunium target elements.

In addition to these main process efforts, various parts of the system have been used for special programs. Highly irradiated plutonium, for example, has been processed in both the solvent extraction and canyon anion exchange systems and finally discharged as oxide from the finishing area. A plutonium isotope mixture containing appreciable Pu-238 has been routinely recovered by solvent batch extraction from the fission product waste stream from enriched uranium processing. Irradiated thorium has been processed in the solvent extraction system to recover U-233 using the THOREX process [5].

2.2.3.2 Material Flowsheet

See Attachment G of this report.

2.2.3.3 Feed Specification

None

2.2.3.4 Production Specification

Same as in Section 2.2.1.3 above.

2.2.3.5 Operating History

F-Canyon went operationally "hot" in November 1954 and H-Canyon in July 1955. Both facilities operated to process natural and/or slightly enriched uranium fuel to recover uranium and plutonium. In February 1957, F-Canyon was shutdown to allow installation of larger Canyon equipment to allow a significant increase in uranium processing capabilities. These modifications were completed by March 1959, and F-Canyon was subsequently restarted. H-Canyon was then shutdown for modifications to allow processing enriched uranium fuel, and was restarted in May 1959.

Plutonium and Uranium processing continued until 1961 when F-Canyon operation was campaigned with that of H-Canyon in order to conserve manpower. Also, from 1964 through 1969, H-Canyon was used for THOREX campaigns for recovery of Uranium-233. In 1966, centrifugal contactors were installed in F-Canyon to replace the "1A" mixer-settler bank to reduce solvent exposure to radiation and subsequent degradation and reduction in performance. Around 1968, F-Canyon began processing Depleted Uranium (DU) targets instead of fuel. In 1972, the Multi-Purpose Processing Facility, or MPPF was completed initially to recover Californium, but actually operated from 1978 to 1980 to process Americium.

PUREX processing continued until 1992 when both canyons were shutdown due to concerns about the ventilation systems reliability to operate in the event of an earthquake. Operations were again curtailed in 1993 due to Conduct of Operations concerns.

By this time, the Savannah River Site mission had changed from that of nuclear materials production to support the weapons complex, to that of post cold war nuclear materials stabilization. The Defense Nuclear Facilities Safety Board Recommendation 94-1 identified categories of materials requiring stabilization via processing through the Canyons.

F-Canyon successfully completed an Operational Readiness Review (ORR) for restart of the Second Plutonium Cycle in January 1995, and an ORR for restart of the rest of PUREX processing (with the exception of Second Uranium Cycle and FA-Line) in January 1996. Stabilization of Plutonium from the remaining DU targets was completed by January 1997, and to date, continues to operate to stabilize Plutonium, primarily from residues produced from past facility operations.

H-Canyon successfully completed an Operational Readiness Review (ORR) for restart of Dissolving and Head End in October 1997, an ORR for restart of First Cycle operation in May 1998, and is completing a Readiness Assessment (RA) for restart of Second Uranium and Second Neptunium Cycles. Stabilization of Uranium from SRS reactor spent fuel continues to date. There is additionally a substantial quantity of Np-237 and Pu-239 stored in H-Canyon that requires future disposition.

2.2.3.6 Current Status

Both Canyons continue to operate to stabilize nuclear materials consistent with the processing capabilities of each canyon, and the priorities defined in the Recommendation 94-1 Implementation Plan. Depleted Uranium solutions produced from operations in F-Canyon are being stored for future disposition, with the Plutonium solutions being transferred to FB-Line for further processing to metal buttons.

Preparations continue for startup of a small vitrification process in MPPF for stabilization of Americium/Curium solutions currently being stored in F-Canyon.

Enriched Uranium, Plutonium, and Neptunium solutions produced from operations in H-Canyon are also being stored for future disposition. The RA for Second Cycle restart is scheduled for completion this year.

2.3 Recycled Uranium Program at SRS

SRS personnel were exploring the ramifications of using recycled uranium as early as January 1958 [9]. It was about this time that the Combined Operations Working Committee began looking into ways to increase the production of Pu-238 in SRS reactors. The Committee began discussing the problems that might be encountered as a result of using previously-irradiated uranium to make fuel for the SRS reactors, as they wanted to utilize the U-236 generated during exposure to increase Np-237 production, in order to speed the production of Pu-238. Use of recycle uranium differed from virgin uranium in three aspects:

a) An increase in total mass of uranium per unit mass of U-235 (the U-235 content of a fuel tube would be kept practically constant)

b) An increase in 'parasitic' absorption of neutrons in the reactor (absorption in the extra, non-fissionable uranium isotopes).

c) An increase in radioactivity.

The increase in radioactivity in the recycle uranium would make the Total Count Instrument (TCI), being used to measure the uranium concentration in U-Al billets useless, because of the increase in specific gamma activity. By March of 1958 efforts were underway to design, develop, and test a replacement instrument for the TCI, which would be unaffected by fission products in the recycle uranium. This effort resulted in the development of an instrument, which used neutron multiplication to measure uranium concentrations in fuel cores. A suitable instrument had been developed by late 1960 and the site proceeded cautiously with plans to utilize recycled uranium in its production reactors [10], [11].

A pilot test was conducted under Test Authorization (TA) 3-529 to process up to 20 kg of recycled uranium to make a preliminary evaluation of the radiation and contamination hazards, as well as, process difficulties associated with the use and handling of recycled uranium. Realizing that the presence of fission products in the recycled material might significantly increase radiation and contamination hazards above those being encountered with virgin uranium, tentative specifications were provided to Union Carbide Nuclear Company (UCNC) in procuring recycled uranium for the pilot test. Those specifications were as follows:

- Maximum gamma activity from fission products shall be 0.05 micro-curies/gram of uranium. This gamma activity was in addition to the approximate 1 micro-curie/gram resulting from the uranium isotopes.
- The alpha activity from plutonium cannot exceed 0.1 micro-curies/gram of uranium.

The maximum radiation exposure of any operating personnel handling uranium meeting the above specifications was estimated to be 6 mr/hr at one foot, with the maximum exposure being encountered at charge weighing and preparation. Since the ability of UCNC to meet the specification had not been demonstrated, provisions were made to receive and process the recycle uranium at SRS only if it could be done safely under Special Work Permits and modified procedures to reduce radiation hazards. These provisions were not utilized since the recycled uranium at UCNC had aged for about 4 months, allowing the fission products to decay sufficiently, such that meeting the required specification was not a problem.

The 20kgs of once-recycled uranium obtained from UCNC were used to manufacture Mark VI-J fuel tubes. These tubes were fabricated in accordance with standard production procedures and processed through normal 300-Area nondestructive tests to provide information on the following:

- Radiation hazards in the storage vaults.
- Radiation and contamination hazards at charge make-up.
- Radiation and contamination hazards during melting, casting, and machining.

- Radiation hazards at other subsequent fabrication steps (i.e., billet assembly and welding, extrusion, etc).
- Process problems with the increased uranium concentration.
- Suitability of the neutron multiplication instrument for determination of core concentration.
- Effects of gamma radiation on the Nuclear Test Gauge (NTG) and gamma scanner results.

Testing protocol required that a sample of recycle uranium from each shipment be analyzed for fission product and Pu content. Extensive Health Physics surveys were also made during the processing of initial shipments of recycle uranium to verify that the Health Physics limits were adequate [12].

The evaluation of radiation and contamination hazards covered by TA 3-529 was accomplished using the first production batch of recycle uranium processed under TA 3-537. The test found that anticipated radiation and contamination hazards were not encountered. The 200-Area process reduced the Pu-alpha to about 0.003 micro-curies/gram of uranium, which was well below the specification of 0.1 micro-curies/gram. The decontamination factor of the UCNC process proved to be large, such that the fission product gamma activity was approximately 0.001 micro-curies/gram of uranium, compared with the specification of 0.05 micro-curies/gram. These large decontamination factors achieved by 200-Area and UCNC were such that Health Physics aspects would not be a problem with recycle uranium, unless the 100-Area cooling period (200 days) was reduced drastically. Health Physics surveys of the Building 321-M process revealed no differences in contamination levels between recycled uranium and regular virgin uranium. The test conclusion and recommendation was to maintain the Pu-alpha and fission product radiation specifications at 0.1 and 0.05 micro-curies/gram respectively for recycle uranium. The conclusion of the test was approved and the recommendation accepted, such that all subsequent receipts of recycle uranium were required to meet the established specifications [13], [14].

Subsequent studies on dose contribution from plutonium and other impurities in uranium waste streams confirm the validity of these early tests. A recent study by Dr. Kenneth W. Crase and Thomas R. La Bone, of the SRS Health Physics Technology Section, Safety and Health Operations Department (ESH-HPT-2000-00040, Dose Contribution from Plutonium and Other Impurities in Uranium in Waste Streams from Savannah River Site Uranium Recovery Facilities) states the following: "We have evaluated dose contributions based on waste stream radioisotope characterizations for the two Savannah River Site (SRS) uranium recovery facilities. These waste streams are considered to contain the maximum concentration of impurities encountered in any SRS facility in which uranium was processed or handled. These waste streams likely also represent a larger concentration of impurities than particular batches of recycled uranium processed, since these facilities processed uranium to remove as many impurities as feasible.

Dose fractions calculated from the radioisotope mix for the SRS uranium recovery facilities indicate that impurities do not contribute a significant fraction of the total dose. For the enriched uranium recovery facility, the total dose fraction due to impurities was less than 8 %, assuming intake parameters that would maximize the internal dose contribution from impurities. For intake parameters that would maximize the internal dose from all radionuclides (including uranium), the impurity dose contribution is much less than 1 %. In the depleted uranium recovery facility, impurities could contribute up to a maximum of 16 % of the total dose, again assuming intake parameters that would maximize the internal dose from impurities. For intake parameters that would maximize the internal dose from all radionuclides (including uranium), the dose contribution from all impurities is much less than 1 %. In none of the cases did any single radioisotope contribute as much as 10 % of the total dose. Even using these conservative assumptions, the results support the SRS internal dosimetry practice of not monitoring SRS uranium workers routinely for plutonium or other actinides.

These results are valid for uranium processed and handled in SRS facilities. They may not be applicable for recycled uranium from SRS that may have been shipped to other nuclear facilities for additional processing or mixing" [15].

2.4 High Potential Worker/Recycle Uranium Contact Activities

Fuel manufacturing activities having the highest potential for Uranium assimilation in Building 321-M were casting charge preparation, casting, U-Al alloy machining, and HEPA filter changes (i. e., those activities where the recycled enriched uranium metal and unclad intermediate U-Al products were processed). Once the cores were assembled into billets and extruded into the clad fuel tubes, there was significantly reduced potential for contamination or assimilation. Charge preparation included receipt, de-packaging, storing, and weighing out quantities of the uranium metal for the casting operation. Casting involved melting (at 800-1200 degrees C) and alloying quantities of uranium metal, U-Al scrap, and aluminum. Machining involved sawing and lathe machining the U-Al alloy to produce billet cores. These activities were conducted in ventilated enclosures with HEPA filtered exhausts, engineered to minimize the contamination/assimilation potential. In addition, operators wore respirators and protective clothing. However, despite these precautions, uranium assimilations did occasionally occur and were detected through the site's bioassay program. Assimilations received focused management attention to identify and correct the causes to prevent recurrences.

There was little contamination/assimilation associated with the Building 313-M slug manufacturing process (i. e., there were no machining operations on the uranium metal, the cores were nickel plated and clad soon after receipt).

Recycle Uranium has been processed through both F-Canyon and H-Canyon. However, radio-chemical processing operations having the greatest potential for worker exposure to recycle uranium contaminants occurred in the FA-Line Facility. Within the canyons, uranium processing is commingled with processing of fission products and other transuranics (i.e. plutonium and neptunium primarily) and appropriate measures have been and are taken to protect against exposure to and monitor for fission products and transuranics.

In the Uranium Oxide Conversion Facility (FA-Line) the potential existed for workers to come into physical contact with recycle uranium. In this facility, liquid uranyl nitrate solution from F-Canyon was concentrated and thermally de-nitrated to an oxide powder (UO_3). This powder was vacuumed (gulped) from the denitrator pots by hand, collected on filters, then transferred to a drum loading facility for storage in 55-gallon drums. Other ancillary powder handling operations also occurred in FA-Line.

The nature of the oxide conversion operations, necessitated that workers handle uranium oxide dust, and work in areas where uranium oxide dust was present. Workers in this facility were routinely monitored for and protected from uranium exposure, as it was the radionuclide presenting the greatest health risk to employees. There were no provisions for radiological control or bioassay programs to prevent or monitor for fission product or transuranic exposures to workers in the facility, as the fission products and transuranics present in the uranium, represented less than 10% of the exposure hazard posed by handling of the uranium itself. In this facility there were no processes through which fission products or transuranics could become concentrated preferentially over uranium.

The areas and activities are described in the following table. While this list may not identify every possible exposure pathway, it does represent those areas and actions that the Site Team, Working Group Team, and Headquarters Team believe to present the highest probability for worker exposure. The Probability for worker exposure is expressed as a number in the "Occupational Exposure Potential" column. This number is the product of three other numbers developed by the Site Teams. Each Site Team reviewed their activity to answer the following questions:

1. How much (high, medium, or low) airborne dust is generated by the activity?

2. What is the radiological hazard (high, medium, or low) of the airborne material generated by the activity? And

3. What is the length of time (long, medium, or short) a worker would be exposed to the airborne materials?

Activities requiring long term exposure to high levels of dust with high radiological activity received the highest score (3x3x3=27); while short duration activities in clean areas received the lowest score (1x1x1=1).

Table 1. Activities with highest Potential for Worker Exposure to Airborne Material

Location	Activity	Time Frame	Potential Constituents of Interest	Occupational Exposure Potential
Bldg. 321-M	Casting Charge Preparation – Charge preparation included receipt, de-packaging, storing, and weighing out quantities of uranium metal for the casting operation. Two workers/ 8-hour shift, 3-shifts/day handled pieces of uranium metal, containing very small amounts of Np, Pu, and Tc. This activity created a minimal airborne and worker exposure potential.	1952-1992	Maximum levels reported in material received on-site* 1.75 ppb Pu 299 ppb Np 2279 ppb Tc	1x1x3=3
Bldg. 321-M	Casting – Involved melting and alloying quantities of uranium metal, U-Al scrap, and aluminum. Furnace operation required 4-6 people/8-hour shift, 3-shifts/day. This activity created medium airborne and high worker exposure potential.	1952-1992-	Maximum levels reported in material received on-site* 1.75 ppb Pu 299 ppb Np	2x1x3=6

			2279 ppb Tc	
Bldg. 321-M	U-Al alloy machining – Machining involved sawing and lathe machining the U-Al alloy to produce billet cores. This activity required 2-4 people/8-hour shift, 3-shifts/day and produced metal shavings and fines, with low airborne and high worker exposure potential.	1952-1992	Maximum levels reported in material received on-site * 1.75 ppb Pu 299 ppb Np 2279 ppb Tc	3x1x3=9
Bldg. 321-M	HEPA Filter change-out – Building exhaust HEPA filters were changed out 6-8 times per year, requiring 2-3 people for each change-out. This activity created high airborne and low worker exposure potential.	1952-1992	Maximum levels reported in material received on-site. * 1.75 ppb Pu 299 ppb Np 2279 ppb Tc	3x1x1=3
F Area A-Line	Facility clean-up – Clean-up of the facility involved removing UO3 dust from floors and equipment each shift. Usually 4-5 people for 1-hour/shift, 3-shifts/day. This activity created high airborne and low worker exposure potential.	1955-1990	Average recorded levels for UO3 Shipped 1.42 ppb Pu 150.6 ppb Np 1.08 ppm Tc **	3x1x1=3
F Area A-Line	Removal of UO3 from denitrator – UO3 powder was vacuumed from the denitrator pots by hand, collected on filters, then transferred to drum loading facility for storage in 55-gallons drums. This activity required 4-5 people/8-hour shift, 3-shifts/day and created high airborne and high worker exposure problems.	1955-1990	Average recorded levels for UO3 Shipped 1.42 ppb Pu 150.6 ppb Np 1.08 ppm Tc **	3x1x3=9

*Constituent data for receipts at SRS utilizes information provided in the Fernald draft report, as no analytical data was available on receipts from plant records. Concentrations of Pu, Np, and Tc are taken from Table F-3-1 of that report. The values chosen are those for normal and enriched shipments of metal which provides the highest level of constituent concentrations shipped to Savannah River Site.

**Tc values are taken from DPST-84-385 as no other analytical data existed for Tc in SRS recycled uranium shipped from the site.

2.5 Reportable Environmental Releases of Recycled Uranium TRU/FP

Uranium releases generally have been associated with the fabrication of reactor fuel and target elements (M Area) and with the chemical processing of spent target and fuel material (F-Area and H-Area).

A review of the uranium releases from SRS facilities to site streams show that 97% of the releases occurred from M-Area operations and the remaining 3% from all other SRS facilities. M-Area operations released about 25 Ci or 96,000 pounds to Tims Branch, a tributary of Upper Three Runs Creek, and most of that uranium remains in the sediments of the Tims Branch system. The uranium concentration at the mouth of Upper Three

Runs Creek is about 0.2 ppb or about 1% of the proposed drinking water concentration guide of 20 ppb. The loss rate of the uranium is extremely low from the Tims Branch system, and initial estimates, based on 6 years of data, indicate a residence time of 2000 years. The uranium concentration in Tims Branch is about 4 ppb (approximately 20% of the proposed drinking water guide of 20 ppb). Uranium concentrations in other SRS streams are at or near background concentrations (0.05 ppb).

Approximately 0.9 Ci of uranium were released to the atmosphere from 1955 through 1996. The dose to the maximally exposed individual at the Site boundary was about 0.4 mrem, and the population dose was 32 person-rem.

The dose from liquid releases of 25 Ci of uranium was 0.5 mrem to the maximally exposed individual, and drinking water doses at the water treatment plants were 0.4 mrem or less. The population dose was 8 person-rem—almost all of it due to drinking water at Beaufort-Jasper and at Port Wentworth. Uranium releases accounted for less than 1 % of the population dose from the Site operations.

For a detailed description of uranium releases to the environment at SRS see WSRC-TR-96-00162, Assessment of Radionuclides in the Savannah River Site Environment-Summary [16].

2.6 Environmental Monitoring and Records

The SRS has been concerned with stewardship of the environment as shown through its policies, procedures, and performance since the pre-construction days of the early 1950s. The first major scientific activity at the site was the determination of the natural levels of radioactivity and the biological condition of the Savannah River. The first task done by the nucleus of what was to become a large environmental monitoring staff, included establishing standard procedures for sampling and analyses at permanently located stations within the site, and as far as 100 miles away from the site. Inventory of about 800 species of flora and fauna at five permanent stations above, along, and below the site boundaries was a second major scientific activity performed at the site and was conducted by the Limnology staff of the Academy of Natural Sciences of Philadelphia.

These early efforts indicate that the utmost importance was placed on protecting the environment and the population from any adverse effects of the SRS operations. It should be noted that at the time the plant was designed and built there were essentially no laws governing discharges into the air or streams. However, even siting of the site was designed to allow space and distance within which to provide barriers and mitigate effects of site operations on the local populace [3].

Through the years, environmental programs have evolved to complement site missions. Policies related to these programs were formalized in recent years in the SRS Environmental Management System Policy, which emphasizes vigilance in protecting human health and ecological or natural resources. SRS is the largest multi-organizational and multi-functional operating nuclear site in the United States to achieve ISO 14001 certification. This certification provides tangible evidence to stakeholders, of SRS commitment to an environmentally safe site, to pollution prevention, to environmental compliance, and to continual improvement.

Notwithstanding all the efforts to protect the environment at SRS, during the years of operation portions of the site have been contaminated. Groundwater beneath an estimated five to 10 percent of the site has been contaminated by industrial solvents, tritium, metals, or other constituents used or generated by operations at SRS. Releases to the environment have been well documented over the years in various environmental reports. Details on SRS environmental contamination and impacts on the surrounding environs can be found in Savannah River Site Environmental Reports, the latest published in 1998, is WSRC-TR-98-00312 [4].

3.0 Recycled Uranium Mass Flow

3.1 Uranium Recycle Description

The SRS primarily received recycled uranium in metal form in its Fabrication Facilities in M-Area. Major suppliers of SRS uranium were facilities at Weldon Springs, Sylvania-Corning Co. (a private concern) Fernald, and Oak Ridge. The material was fashioned into fuel and target elements, irradiated, subjected to chemical separation to recover product and to recover usable uranium. This uranium usually in the form of Uranium Trioxide (UO3) or Uranyl Nitrate (UNH) was shipped to Fernald, Y-12, Paducah, and other minor sites to be placed back into the recycle stream. Portions of the uranium were lost to waste streams in both the fabrication and separation processes and remain on the Site in settling basins and waste storage facilities. The bulk of the UO3 produced at the Site (about 20,000 MTU) remains in storage at SRS. A host of other sources including other government sites, colleges and universities, and foreign entities, also shipped small amounts of uranium to SRS. Details of shipments and receipts of recycle uranium are provided in the Sections below.

3.2 Uranium Receipts

A search of Material Control and Accountability Records indicates that from the time SRS started to handle uranium to March 31, 1999 some 54,544 metric tons of uranium were received at the site. The bulk of this material (45,342 MTUs) was received as uranium metal from Fernald. Another 180.8 MTUs of uranium metal came from the Y-12 Plant at Oak Ridge. The remainder of the uranium receipts were in the form of oxides, 864 MTUs from Fernald, 4.2 MTUs from Y-12, 5 MTUs from Paducah, and 14 MTUs from the Oak Ridge K-25 Plant. SRS receipts from other minor sites totaled 8, 134 MTUs. The receipts from the major shipping sites are shown annually by shipping site and material form in Attachment A.

3.3 Uranium Shipments

A search of Material Control and Accountability Records indicates that from the time SRS started to handle uranium to March 31, 1999 some 31,355 MTUs were shipped from the SRS to other sites within the DOE Complex. The bulk of these shipments went to K-25 (10,290 MTUs of oxide from 1955 to 1999), Paducah (9,257 MTUs of oxide from 1955 to 1999), and Fernald (8962 MTUs of oxide and scrap). Another 64.3 MTUs of oxide and scrap went to Y-12 along with 91 MTUs of solution. The remainder of the shipments was 74 MTUs of solution shipped to Fernald and 2, 617 MTUs shipped to a host of minor sites. The shipments to the major sites are shown annually by receiving site and material form in Attachment B.